

# Fractional charges in pyrochlore lattices

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**Abstract.** A pyrochlore lattice is considered where the average electron number of electrons per site is half-integer, concentrating on the case of exactly half an electron per site. Strong on-site repulsions are assumed, so that all sites are either empty or singly occupied. When there are in addition strong nearest-neighbour repulsions, a tetrahedron rule comes into effect, as previously suggested for magnetite. We show that in this case, there exist excitations with fractional charge  $\pm e/2$ . These are intimately connected with the high degeneracy of the ground state in the absence of kinetic energy terms. When an additional electron is inserted into the system, it decays into two point like excitations with charge  $-e/2$ , connected by a Heisenberg spin chain which carries the electron's spin.

**Keywords:** pyrochlore lattice, fractional charge, heavy fermions

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## 1 Introduction

The experimental observation of heavy-fermion behaviour of  $\text{LiV}_2\text{O}_4$  [1, 2, 3] has drawn attention to pyrochlore lattice systems with half-integer valency of the involved ions. For example, the average valency of V in  $\text{LiV}_2\text{O}_4$  is +3.5, and the same is true for Ti in  $\text{LiTi}_2\text{O}_4$ . Therefore, the average 3d electron number is  $d^{1.5}$  in the first case, and  $d^{0.5}$  in the second one. From LDA band-structure calculations [4, 5, 6, 7] it is known that the conduction bands have 3d- $t_{2g}$  character, and are well separated from higher energy valence electron states. However the LDA effective electron mass found for  $\text{LiV}_2\text{O}_4$  is a factor of twenty-five smaller than the quasiparticle mass inferred from specific heat and spin susceptibility data. This is a sign of strong electronic correlations. On-site Hubbard  $U$  interactions alone are not sufficient to explain the large measured quasiparticle mass. They merely reduce the atomic configurations of the V ions to  $3d^1$  and  $3d^2$ , i. e. , they exclude  $3d^0$ ,  $3d^3$  configurations, etc. In order to obtain a sufficiently high density of low-energy excitations one must therefore include correlations between neighbouring sites. The nearest-neighbour interactions are minimised if, for each of the corner sharing tetrahedra which make up the pyrochlore lattice, there are two  $3d^1$  and two  $3d^2$  configurations. This so-called “tetrahedron rule”, which is implicit in Verwey’s treatment of the metal-insulator transition in the spinel  $\text{Fe}_3\text{O}_4$  [8], was first stated explicitly by Anderson [9]. The number of configurations which obey the

tetrahedron rule grows exponentially with the number of tetrahedra, so in the absence of any perturbation (such as the electrons' kinetic energy) which selects between these states, the ground state of pyrochlore systems with nearest-neighbour interactions and half-integer valency is extremely degenerate. An important property of these degenerate ground-state configurations is that each of them consists of chains of  $3d^1$  and  $3d^2$  sites in the case of  $\text{LiV}_2\text{O}_4$  [10], and of empty  $3d^0$  and  $3d^1$  sites in the case of  $\text{LiTi}_2\text{O}_4$ . Spin excitations in those chains can give rise to a linear low temperature specific heat  $C(T)=\gamma T$  with a large  $\gamma$  coefficient [10]. Because of the special features of one-dimensional systems, these spin excitations can be described by either bosons or fermions [11].

In this paper we set to explore the effects of a small kinetic energy term on a system obeying the tetrahedron rule. For simplicity we shall limit our discussion to the case of an average  $3d^{0.5}$  count, i. e. , to states with one electron for every two lattice sites. We note that the calculations are not intended to apply to  $\text{LiTi}_2\text{O}_4$ , for which no heavy-fermion behaviour has been observed, but should instead be considered as having model character. They are intended to demonstrate special features of excited states in charge-frustrated lattices. In order to be better able to visualize states obeying the tetrahedron rule, we further simplify the problem by considering a two-dimensional checkerboard lattice instead of the original three-dimensional pyrochlore lattice. This is essentially a special projection of the pyrochlore lattice onto a plane, and all of the results we discuss apply equally to three-dimensional pyrochlore systems.

## 2 Model Hamiltonian

For a pyrochlore lattice with  $t_{2g}$  electrons, the full model Hamiltonian is of the form [13]

$$\begin{aligned} \mathcal{H} = & \sum_{\substack{\nu=1 \\ \mathbf{k}, \sigma}}^{12} \epsilon_{\nu}(\mathbf{k}) a_{\mathbf{k}\nu\sigma}^{\dagger} a_{\mathbf{k}\nu\sigma} + (U - 2J) \sum_{ia} n_{a\uparrow}(i) n_{a\downarrow}(i) \\ & + \frac{1}{2} \left( U - \frac{J}{2} \right) \sum_{\substack{a \neq a' \\ i}} n_a(i) n_{a'}(i) - J \sum_{\substack{a \neq a' \\ i}} \mathbf{s}_a(i) \cdot \mathbf{s}_{a'}(i) \\ & + \bar{J} \sum_{\langle ij \rangle} \mathbf{S}(i) \cdot \mathbf{S}(j) + V \sum_{\langle ij \rangle} n(i) n(j) \quad . \end{aligned} \quad (1)$$

The first term describes the kinetic energy of the  $t_{2g}$  electrons which form twelve bands with index  $\nu$  because of the four sites per unit cell. The following three terms are due to Coulomb repulsions of d electrons on a transition-metal ion site  $i$  occupying orbitals  $a$  and  $a'$ . Here  $U$  is the direct and  $J$  the exchange part of the on-site Coulomb interaction; the latter enforces Hund's first rule when several d electrons occupy the same site. We shall assume that  $U$  is large enough to ensure that each ion fluctuates between two valencies only. The remaining two terms describe the exchange  $\bar{J}$  and direct Coulomb  $V$  interactions between neighbouring sites. These interactions are *frustrated* by virtue of the geometry of the pyrochlore lattice, and this is an essential

prerequisite of our theory. In particular, it is the nearest neighbour repulsion  $V > 0$  which singles out states obeying the tetrahedron rule.

In order to first discuss excitations associated with charge degrees of freedom, we begin by considering the reduced Hamiltonian

$$\mathcal{H}_0 = -t \sum_{\langle ij \rangle} \{ f^\dagger(i) f(j) + h. c. \} + V \sum_{\langle ij \rangle} n^f(i) n^f(j) \quad (2)$$

where  $n^f(i) = f^\dagger(i) f(i)$ . This refers to a system with an average d-electron count of 0.5 and one orbital per site. Because of the assumed large value of  $U$ , only empty ( $d^0$ ) and singly occupied ( $d^1$ ) sites are considered. Spinless fermions are assumed, which corresponds to a full spin polarisation of the electrons. Charge conservation requires the subsidiary condition

$$\sum_i n^f(i) = N/2 \quad (3)$$

for a system with  $N$  lattice sites, while in the limit  $V \rightarrow \infty$  the tetrahedron rule requires that

$$\sum_{\nu=1}^4 \left( n^f(i_\nu) - \frac{1}{2} \right) = 0 \quad (4)$$

where  $\nu$  denotes the four different sites within *any* given tetrahedron  $i$ . In one dimension, this Hamiltonian can be mapped onto an anisotropic Heisenberg model, and can be solved exactly [12]. In this case one finds a gap-less metallic excitation spectrum for  $V < 2t$ , and a gapped insulating state for larger values of  $V$ .

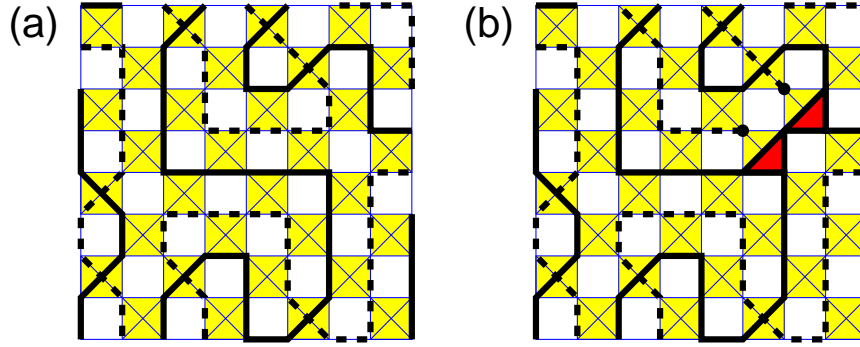
For a pyrochlore lattice no such exact results are available. However, we expect that the parameter range for the two phases remains qualitatively unchanged, since the increased number of nearest neighbours enters the hopping and interaction terms in the same way.

### 3 Fractional charge

We consider a two-dimensional checkerboard lattice consisting of  $N$  sites with a configuration  $|\Phi_I\rangle$  representing one of the degenerate ground states, i. e. , a configuration in which the tetrahedron rule is satisfied (for an example see Fig. 1a). It is known that, for the 2D checkerboard lattice, the number of these configurations scales as  $(4/3)^{3N/4}$  [17]. The wavefunction of  $|\Phi_I\rangle$  is of the form

$$\Phi_I \left( 1, \dots, \frac{N}{2} \right) = \mathcal{A} \prod_{\nu} \phi(\nu(1), \dots, \nu(N_{\nu})) \quad (5)$$

where  $\sum_{\nu} N_{\nu} = N/2$ , and the operator  $\mathcal{A}$  antisymmetrizes the wavefunction. Here  $\phi(1, \dots, M)$  is the exact ground-state wavefunction of a Heisenberg chain with an even number  $M$  of sites. The chains have to be arranged in such a way that the tetrahedron rule is satisfied by every cross-linked square of the lattice.



**Fig. 1** Checkerboard lattice, the thin lines indicate hopping and interactions connected sites. Thick lines connect sites in a  $d^1$  configuration while dotted lines connect empty ( $d^0$ ) sites. (a) example of a fraction of the lattice in which the tetrahedron rule is obeyed. (b) the sample as (a) but with an electron added ( $d^0 \rightarrow d^1$ ). Dots indicate the end of the chain.

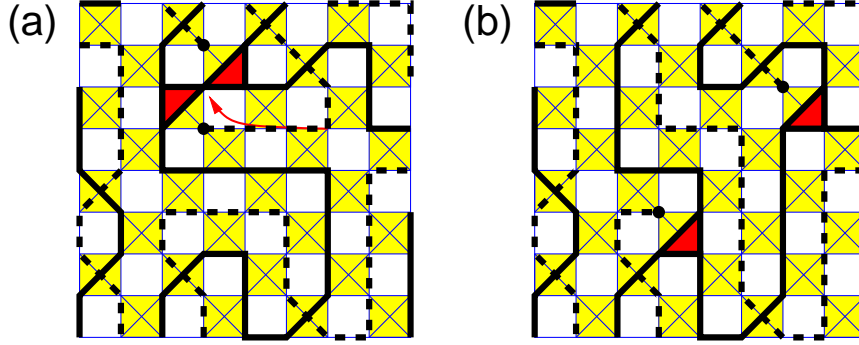
When a charge  $-e$  is added to an empty site  $j$  we are left with two corner-sharing squares with three  $d^1$  sites each. This is illustrated in Fig. 1b. The corresponding state is  $|j\rangle = f^\dagger(j) |\Phi_I\rangle$ . The kinetic energy term now permits the five electrons within these two squares to move. However, the motion has to occur in such a way that the number of squares with three  $d^1$  sites is conserved. The energy barrier to generating states with additional violation of the tetrahedron rule is  $V$ , which has already been assumed to be sufficiently large to prevent this from happening. And, critically, there is no way to “undo” the violation of the tetrahedron rule geometrically, simply by moving electrons around the lattice. The number of tetrahedra (squares) in which the tetrahedron rule is violated is therefore a topological invariant of the states which we consider.

If the added electron moves along the chain of empty sites in which it was inserted, it retains its integrity and the state  $|j\rangle$  transforms successively into states  $|n\rangle = f^\dagger(n)f(j) |j\rangle$ . However, if we instead allow one of the four neighbouring electrons belonging to chains of occupied sites to move, the electron immediately breaks into two disjoint pieces. These carry fractional electric charge  $-e/2$  (note that every electron is shared by two squares and so contributes a charge  $-e/2$  to each of them). These two cases are indicated in Figs. 2a,b.

Energy and momentum (as well as topological charge) must be conserved by these decay processes. Therefore if we were to associate a momentum  $\mathbf{k}$  and energy  $E(\mathbf{k})$  with the electron which we inserted, this must now be shared between the fractionally charged particles into which it has decayed

$$E(\mathbf{k}) = 4V + \epsilon(\mathbf{k}_1) + \epsilon(\mathbf{k}_2) \quad (6)$$

where  $\epsilon(\mathbf{k})$  is the dispersion of the fractional charge and  $\mathbf{k}_1 + \mathbf{k}_2 = \mathbf{k}$ . The situation resembles that of spin excitations in a Heisenberg chain. There, a spin 1 spin-flip excitation can decay into two spin 1/2 domain walls (spinons), with the result that for each value of  $k$  there is a continuum of spin excitations. Clearly, a metal which



**Fig. 2** (a) The added electron has moved in four steps along a chain of empty sites. It remains an entity. (b) An electron from lower triangle in Fig. 1b has moved along the diagonal. As a consequence the excitation has decayed into two with a fractional charge of  $-e/2$  each.

has a charge excitation spectrum with low energy contributions of the form of Eq. (6) *cannot* be a conventional Fermi liquid.

We can obtain some feeling for the form of  $\epsilon(\mathbf{k})$  by modelling the fractional charge as a free particle on a checkerboard lattice with a reduced bandwidth due to the hopping restrictions imposed by the tetrahedron rule. In the case of hopping between nearest-neighbour sites only, this would be a band with dispersion  $\epsilon(\mathbf{k}) = -t[\cos(k_x a) + \cos(k_y a)]$ , where  $a$  is the lattice constant. Without the hopping restrictions the prefactor would be twice as large, and the band two-fold degenerate; if the diagonal hopping elements were also restored one of the two bands would become dispersionless. The corresponding free electron dispersion for the pyrochlore lattice, including flat bands, is given in [14].

Physically, the fractionally charged excitations in our model result from a back-flow of charge. When a fractional charge travels along a given path, it leaves behind it a “wake” of squares (tetrahedra in the case of a pyrochlore lattice) which still obey the tetrahedron rule, but whose charge configuration has been modified. Motion constrained by the tetrahedron rules occurs such that each “hopping” event of an electron is accompanied by a net back-flow of a charge  $-e/2$ . A fractional charge of  $-e/2$  is therefore carried by the moving particles. We want to stress that all of these arguments about fragmentation of charge depend only on the geometry of the lattice (more precisely, on the geometry of the repulsions  $V$ ), and not on its dimensionality or details of the band structure. Everything which has been demonstrated here for the checkerboard lattice therefore also applies to the three-dimensional pyrochlore lattice.

#### 4 Spin of fractional charges

When an electron is added to an empty site, a spin  $1/2$  is added to the system. In order to gain some insight into how this spin is distributed when the electron decays

into two fractional charges, we now restore a spin degree of freedom to each  $d^1$  site and consider the model

$$\mathcal{H} = \mathcal{P} \left[ -t \sum_{\langle ij \rangle \alpha} \{ d_{\alpha}^{\dagger}(i) d_{\alpha}(j) + h. c. \} \right] \mathcal{P} + V \sum_{\langle ij \rangle} n^d(i) n^d(j) + \bar{J} \sum_{\langle ij \rangle} \mathbf{S}(i) \cdot \mathbf{S}(j) \quad (7)$$

where  $\alpha = \{\uparrow, \downarrow\}$  is the electron spin index,

$$n^d(i) = \sum_{\alpha} d_{\alpha}^{\dagger}(i) d_{\alpha}(i) \quad , \quad \mathbf{S}(i) = \frac{1}{2} \sum_{\alpha\beta} d_{\alpha}^{\dagger}(i) \vec{\sigma}_{\alpha\beta} d_{\beta}(i) \quad ,$$

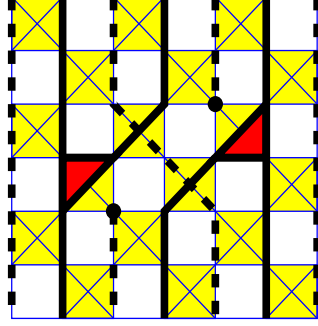
and

$$\mathcal{P} = \prod_i [1 - n_{\uparrow}^d(i) n_{\downarrow}^d(i)] \quad (8)$$

is a (Gutzwiller) projection operator ensuring that no site is doubly occupied. At exact quarter filling, all configurations which obey the tetrahedron rule are composed of randomly distributed Heisenberg spin chains, which, for periodic boundary conditions, have even length. Only exchange interactions then come into play, and these select a singlet ground state.

When an electron is added to one of these states it either connects two adjacent spin chains, as illustrated in Fig. 1b, or two neighbouring sections of a single closed spin-chain loop. Let us suppose for a moment that  $t = 0$ , and the electron is fixed at this site. From density-matrix renormalisation group (DMRG) calculations it is known [15] that a spin 1/2 coupled symmetrically to a single Heisenberg chain has a ground state which is a Kramers doublet, and a Curie-like susceptibility. We expect the same to be true for a spin 1/2 coupled symmetrically to two Heisenberg chains as in Fig. 1b. Again the frustration of the lattice plays a key role in determining the nature the states, but this time it is the spin interactions which are frustrated.

If we now restore the kinetic energy term, we once again have two possible cases for electronic motion. When the electron moves along a chain of empty sites, its spin interactions are at every step frustrated, and so it can carry its spin 1/2 intact as it moves. But as soon as the electron decays into two fractional charges, its spin is no longer localised at one point. In most cases, the insertion of an electron will connect two different spin chains. The decay of the electron converts a single spin 1/2 site into a spin chain with an odd number of spin 1/2 sites coupled symmetrically to two Heisenberg chains with an even number of sites. Since the total number of sites involved is odd, the ground state remains a Kramers doublet, as required. But it is no longer possible to uniquely assign this spin 1/2 degree of freedom to a single site. This situation is illustrated in Fig. 3. For better visualisation we have chosen here a special configuration in which in the absence of the added electron all chains have a regular charge order. This is necessary only to be able to illustrate the effect using a small segment of the checkerboard lattice; global charge order is *not* required.



**Fig. 3** A configuration with periodic boundary conditions which demonstrates that two Heisenberg chains are connected by a third one consisting of an odd number of sites.

## 5 Quantum fluctuations

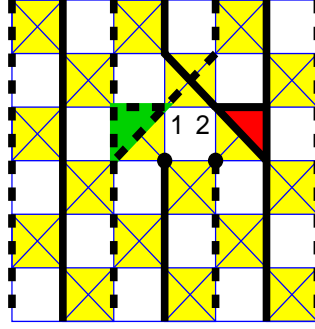
In the arguments so far presented we have considered the limit  $t/V \rightarrow 0$  for which repulsive interaction between electrons on neighbouring sites can be replaced with a simple “tetrahedron rule”. If we relax this constraint to the extent of considering finite (but very large)  $V$ , quantum fluctuations about the many degenerate states obeying the tetrahedron rule can themselves single out a ground state. The problem then falls into the general class of order from disorder effects, widely studied in the context of frustrated magnetic insulators, but largely unexplored for metals.

Here we briefly discuss two aspects of these quantum fluctuations; the fact that they also contain fractional charges, and their relevance for the metal–insulator transition at (smaller) finite  $V$ . We start from one of the degenerate ground states  $|\Phi_I\rangle$  which obey the tetrahedron rule, and consider to lowest order the effect of the kinetic energy. At  $\mathcal{O}(t/V)$  this mixes into the wavefunction virtual states in which two squares (tetrahedra) violate the tetrahedron rule at the net cost of an interaction energy of  $V$ . However overall charge must be conserved, so in this case one square contains three  $d^1$  sites, while the other has three  $d^0$  sites. This is illustrated for the checkerboard lattice in Fig. 4. If this virtual excitation lives long enough for the contributing electrons to hop to neighbouring sites it, just like an added electron, will decay into two fractional charges. In this case the two pieces of the excitation must carry the opposite sign of electrical charge. We have already assigned a charge  $-e/2$  to the square containing three  $d^1$  sites; the square containing three  $d^0$  sites carries a net charge  $e/2$ , and the number of squares violating the tetrahedron rule must remain unchanged, once the vacuum fluctuation has been created.

The energy associated with such a vacuum fluctuation is then

$$\Delta E_{\text{vac}} = V + \epsilon(\mathbf{k}) + \bar{\epsilon}(-\mathbf{k}) \quad . \quad (9)$$

the potential energy lowered by the kinetic energy of the two defects (here we differentiate between the kinetic energy  $\epsilon(\mathbf{k})$  of the defect associated with the three  $d^1$  sites, and the  $\bar{\epsilon}(\mathbf{k})$  of a square containing three  $d^0$  sites). If the net energy of *any*



**Fig. 4** Vacuum fluctuation due to an electron hopping from 1 to 2. Two charges  $\pm e$  are generated which can propagate freely. Note that the Heisenberg chains involve even numbers of sites.

vacuum fluctuation is negative, the tetrahedron rule will break down, and at least in the crudest approximation, a metal–insulator transition will take place. Numerically, for spinless fermions on a checkerboard lattice (of size eight to sixteen squares, with periodic boundary conditions), we find that this occurs for  $V \approx 7t$ .

The structure of the vacuum fluctuation is such that a spin  $1/2$  chain with an *even* number of sites is linked symmetrically to a Heisenberg ring (see Fig. 4), implying a singlet ground state. Importantly, the  $+e/2$  “tail” of the vacuum fluctuation (the tetrahedron with only one  $d^1$  site), can annihilate with one of the fractional charges  $-e/2$  produced by the fragmentation of an added electron. This makes it possible to locally reassemble the added electron anywhere in the lattice. Furthermore, fluctuations can recombine in such a way that they create the objects associated with an extra electron and one hole (the latter being an open chain of spins). Therefore we expect a balance between fractional charges  $\pm e/2$  and full charges  $\pm e$  when electrons (holes) are added to the system, or even in the metallic state.

While the dynamics of the defect is relatively straightforward in the case of spinless fermions, the introduction of spin greatly complicates matters. The kinetic energy term conserves the spin of the mobile electrons, so the motion of a defect is accompanied by drastic rearrangements of the spin in the Heisenberg chains connected by hopping events. We therefore speculate that at energies smaller than the typical scale  $J$  of spin excitations, the effective hopping of fractional charges may be reduced relative to the (coherent) motion of electrons in the channels provided by empty sites.

## 6 Conclusions

In this paper we have demonstrated that excitations with fractional charge  $\pm e/2$  can occur in a pyrochlore lattice with strong short-range Coulomb repulsions. They appear in pairs with a delocalized spin  $1/2$  (electron) or spin 0 (vacuum fluctuation). Fractionally charged excitations were first proposed in the context of heavily doped



trans-polyacetylene by Wu and Schrieffer [16], where they are linked to a commensurate superstructure of the polymer chain. They have also famously been invoked in a ground-breaking paper by Laughlin [18], in connection with the fractional quantum Hall effect. However, polyacetylene is a one-dimensional system, and the quantum Hall effect is a phenomenon occurring in two-dimensions and high magnetic field. To the best of our knowledge the present case provides the first example of a microscopic model supporting fractional charge in three dimensions.

In polyacetylene and related one-dimensional model systems, the prerequisite for the fractionalisation of charge is the existence of degenerate vacua. The crucial prerequisite found here for the appearance of fractional charges is the highly degenerate ground state of the pyrochlore lattice in the absence of kinetic energy terms. Just like a moving soliton in polyacetylene, which transforms one of the two ground-state configurations into the other, a moving fractional charge in a pyrochlore lattice links different degenerate configurations of the system. Furthermore, because at finite  $V$  there exist a density of order  $(t/V)^2$  of vacuum fluctuations which can recombine with fractional charges, the model also supports some admixture of excitations carrying the full electronic charge  $-e$ .

The above considerations apply primarily to pyrochlore insulators, when doped with electrons or holes. An example of such a system is magnetite ( $\text{Fe}_3\text{O}_4$ ), an insulating spinel oxide, whose B sites form a pyrochlore lattice of  $\text{Fe}^{2+}$  and  $\text{Fe}^{3+}$  ions. We note, however, that spinel oxides are prone to structural phase transitions which lift the huge degeneracy of the states obeying the tetrahedron rule. The notable exception to this pattern is  $\text{LiV}_2\text{O}_4$ . Longer-range Coulomb repulsions will also act to reduce the degeneracy. All of these effects will make it harder for fractional charges to propagate freely.

Obviously, many unsolved problems remain. For example, the calculation of thermodynamic and transport properties of the model, or the effect of an applied magnetic field, have yet to be attempted. It would also be worthwhile to search for other lattice geometries in which fractional charges could occur. The present communication can only point to these problems and does not pretend to provide an answer. It should, however, be considered as a first step in that direction.

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